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13. ABSTRACT (Maximum 200 words)

An ONR sponsored workshop titled "Surface Dynamical Growth Processes" was held at Vanderbilt University, Nashville, Tennessee, on October 28-29, 1993 in conjunction with a Free-Electron Laser Program Review. The workshop was sponsored by T. L. Reinecke, N. H. Tolk and S. C. Ying. Leading scientists involved in materials growth research gave presentations on significant growth issues. The presentations led to lively discussions which brought the present state of experimental and theoretical knowledge into sharp focus.

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ONR FREE-ELECTRON LASER PROGRAM ON MATERIALS

October 27, 1993

Chairperson: N. H. Tolk

SURFACE DYNAMICAL GROWTH PROCESSES

October 28-29, 1993

Chairpersons: T. L. Reinecke, N. H. Tolk and S. C. Ying

Vanderbilt University, Nashville TN, USA

Local Committee for both meetings:

A. V. Barnes, Chairman
R. G. Albridge
J. L. Allen
J. T. Mckinley
G. Mensing
S. Vijayalakshmi
B. Stanley, Secretary

ONR Free-Electron Laser
Program on Materials

October 27, 1993

Vanderbilt University

Nashville Tennessee

Table of Contents

Materials Program

October 27, Wednesday

8:00 - 8:30 AM		Registration
8:30 - 8:45 AM		Welcome

SESSION I   **Chairperson: J. L. Davidson**

8:45 - 9:05 AM	**S. C. Ying**	"Diffusive Motion of Molecules"
9:05 - 9:25 AM	**S. Feng**	"Photon Migration in the Presence of A Single Defect: A Perturbation Analysis"
9:25 - 9:40 AM	**C. J. Stanton**	"Transient Photocurrent and Transient Localization of Carriers Under Ultrafast Optical Excitation"
9:40 - 9:55 AM	**J. G. Fujimoto**	"Femtosecond Carrier Dynamics in InGaAs/AlGaAs Strained Layer Single Quantum Well Diode Lasers"
9:55 - 10:35 AM	**M. D. Fayer**	"Picosecond Vibrational Photon Echo Experiments in Liquids and Glasses"
10:35 - 11:00 AM	*** Coffee Break ***	

SESSION II   **Chairperson: R. F. Haglund**

11:00 - 11:20 AM	**P. M. Fauchet**	"Pump-probe and Second Harmonic Generation Measurements in Semiconductors"
11:20 - 11:40 AM	**N. H. Tolk**	"FEL Semiconductor Spectroscopy of Heterojunctions"
11:40 - 12:00 PM	**G. Margaritondo**	"FEL Two-Photon Spectroscopy of Semiconductors"
12:00 - 12:40 PM	**S. J. Allen**	"Probing Terahertz Dynamics in Semiconductor Nanostructures with the UCSB Free-electron Lasers"
12:40 - 2:10 P.M.	*** *Lunch* ***	

Materials Program (continued)

SESSION III	Chairperson:	G. S. Edwards
2:10 - 2:30 P.M.	**B. D. McCombe**	"Saturation Spectroscopy of Free Carriers in Quasi-Two-Dimensional Systems"
2:30 - 3:10 PM	**P. C. Taylor**	"Application of Free Electron Laser Technology to the Study of Energy Transfer in Disordered Solids"
3:10 - 3:30 PM	**N. J. Halas**	"Fullerene Solids: From Properties to Practical Applications"
3:30 - 3:50 PM	**B. Garrison**	"Simulations of Laser Drive Dynamics in Materials and Surfaces"
3:50 - 4:20 PM		***** Coffee Break *****

SESSION IV	Chairperson:	A. V. Barnes
4:20 - 4:40 PM	**S. M. George**	"Distance Dependence of Electronic Energy Transfer Between Donor and Acceptor Adlayers: p-Terphenyl and 9,10-Diphenylanthracene"
4:40 - 5:00 PM	**J. D. Simon**	"Ultrafast Dynamics in Chemical and Biochemical Systems"
5:00 - 5:20 PM	**J. H. Weare**	"Parallel Computers in Large Scale "First Principle" Simulations of Complex Materials"
6:00 - 8:00 PM		***** Reception ***** Registration desk will be open

Materials Abstracts in Order of Presentation

Diffusive Motion of Molecules

S. C. Ying
Brown University

The diffusive motion of adsorbed atoms or molecules is usually studied (both theoretically and experimentally) as a separate subject from that of the vibrational motion near the minima of the adsorption potential. This is certainly well justified at low temperatures when the time scale of the vibrational motion is much shorter than that of activated or quantum diffusion. At higher temperatures, the two time scales may be comparable and it is important to treat the two aspects of the motion in a unified way. Starting with the harmonic description of the frustrated translational mode of the adsorbed atom or molecule, the anharmonicity of the adsorption potential become increasingly important as the temperature increases. This strong anharmonicity leads to a shift and broadening of the frustrated translation mode in addition to those induced through coupling to substrate phonons. At yet higher temperatures, the diffusive motion become important and hence the anharmonicity of the adsorption potential becomes essential and cannot be treated in any perturbative fashion. In this regime the frustrated translational mode gradually becomes overdamped.

We have pursued a unified treatment of the vibrational and diffusive motion of an adatom through the study of the finite frequency velocity auto-correlation function. The theory is based on the Mori projection operator formalism and includes the full effect of anharmonicity of the adsorption potential non-perturbatively as well as the coupling to the substrate phonons. As examples of application of the theory, we will discuss topics such as kinetic coefficients near a structural phase transition, classical to quantum crossover and quasi-elastic peak in scattering experiments.

Photon Migration in the Presence of a Single Defect: a Perturbation Analysis

Shechao Feng and Fan-An Zeng
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Los Angeles, California 90024-1547

Britton Chance
Department of Biochemistry and Biophysics
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We have developed a perturbation theory to study the sensitivity of diffusive photon flux to the addition of a small spherical defect object in multiple scattering media such as human tissue. As a first simple application of our perturbation method, we derive analytically the photon migration path distributions, and the shapes of the so called "banana" regions in which the photon migration paths are concentrated. We then derive analytically the sensitivity of detected photon flux densities to the inclusion of small spherical defects in the multiple scattering medium, for both single source and two-source configurations, at both steady state (DC) and frequency modulation conditions, and compare the results with Monte Carlo simulations.

Transient Photocurrent and Transient Localization of Carriers under Ultrafast Optical Excitation

A. V. Kuznetzov, and C. J. Stanton

Department of Physics
University of Florida
Gainesville, FL 32611

Recent experiments have shown that the ultrafast optical excitation of semiconductors in a DC electric field gives rise to a photocurrent that varies rapidly in time. This current acts as a source for dipole electromagnetic radiation in the form of femtosecond far-infrared pulses. The theoretical understanding of the carrier transport during and immediately after the excitation and the mechanisms responsible for the THZ radiation however, is still incomplete. Theories have attributed the radiation either to "current surges" (particle current due to the photoexcited carriers in the depletion field) or "optical rectification" (displacement current caused by the polarization of the photoexcited electron-hole pairs in a dc electric field). We present a theory which consistently treats *both* transport and displacement contributions to the total photocurrent. Our results indicate that both mechanisms are needed. We demonstrate that ultrafast excitation creates carriers in transiently localized states that are polarized in the DC field and later evolve into extended stationary eigenstates causing transport current in the process. The resulting photocurrent is determined by both transport and displacement components and exhibits nontrivial dependence on the DC field, excitation duration and detuning.

Femtosecond Carrier Dynamics in InGaAs/AlGaAs Strained Layer Single Quantum Well Diode Lasers

J. G. Fujimoto and C. K. Sun

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H. E. Choi and C. A. Wang

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G. D. Sanders and C. J. Stanton

Department of Physics
University of Florida
Gainesville, FL 32611

Nonlinear gain and transient carrier dynamics play an important role in laser linewidth, modulation bandwidth, amplification, and ultrashort pulse generation. We report the first studies of carrier dynamics in InGaAs/AlGaAs strained-layer single-quantum-well (SQW) diode lasers using a novel multiple-wavelength pump probe technique. The use of separate wavelengths for the pump and probe pulses permits a more direct measurement of transient carrier dynamics than is possible using conventional single-wavelength pump probe techniques. Transient behavior is governed by carrier temperature dynamics from free carrier absorption and stimulated transitions. Different processes dominate the transient response depending upon the pump energy relative to the transparency energy and the average carrier energy.

Picosecond Vibrational Photon Echo Experiments in Liquids and Glasses

Michael D. Fayer

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Stanford University, Stanford, CA 94305

Picosecond vibrational photon echo experiments in liquids and glasses are described. The experiments were conducted using the Stanford Super Conducting Free Electron Laser. The CO stretching mode at 5.06 μm of tungsten hexacarbonyl in 2-methyl THF was examined. The experiments were performed as a function of temperature and observed vibrational dephasing through the glass transition. The pure vibrational dephasing exhibits a power law temperature dependence at low temperature that becomes exponentially activated near the glass transition temperature. In addition, pump-probe experiments are used to measure vibrational life times as a function of temperature from room temperature to 16 K. This is the first study of the temperature dependence of a vibration that spans a range including the liquid, super-cooled liquid, and glass. In the liquid, the decay is a bi-exponential. It suddenly changes to a single exponential when the liquid becomes super cooled. Possible mechanisms for the temperature dependence of the vibrational dephasing and the vibrational life time are discussed.

Pump-Probe and Second Harmonic Generation Measurements in Semiconductors

Philippe M. Fauchet

Department of Electrical Engineering &
Laboratory for Laser Energetics &
Center for Optoelectronics and Imaging
University of Rochester, Rochester NY 14627

I will discuss the results of two types of experiments we have performed with a free electron laser in the past 4 years. In one type of experiment, we completed the first two-color pump-probe measurements using a FEL tuned to 1.5 μm, The experiment revealed carrier recombination and trapping in amorphous silicon-germanium. In the other type of experiments, we have measured second harmonic generation in p-type gallium arsenide quantum wells. The measurements have been performed using an FEL tuned in the 3.5 to 5.5 μm region.

Free-Electron Laser Semiconductor Spectroscopy of Heterojunctions†

N. H. Tolk*

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Vanderbilt University, Nashville, TN 37235

A materials science collaboration between the Ecole Polytechnique Federale in Lausanne and Vanderbilt University have produced interesting new results using the Vanderbilt Free-Electron Laser, the most powerful tunable FEL in the world (2-10 μm, extendible to 1 μm by frequency doubling). Heterojunction band discontinuity measurements of unprecedented accuracy have been carried out. FEL internal photoemission (FEL-IPE) measurements of conduction band discontinuities at GaAs-GaAlAs, and Ge-GaAs interfaces are presently accurate to better than ±5 meV. We emphasize that FEL-IPE is a *direct* measurement of the discontinuity based on the photocurrent threshold that occurs when the photon energy exceeds the conduction band discontinuity. Since FEL-IPE uses subband gap photons, it can be applied to device-quality interfaces deep beneath a surface. The effective interface band gap, which can vary from the bulk gap due to microdiffusion, is also measurable. Systematic use of this technique over a wide range of semiconductor interfaces promises to resolve long-standing disagreements between competing semiconductor band-lineup theories, and provide important information on the influence of defects and Δ-layers.

Materials Science on the Vanderbilt FEL: First Results and Ongoing Programs†

G. Margaritondo*

*Ecole Polytechnique Fédérale,
Ch-1015 Lausanne, Switzerland.*

The Ecole Polytechnique Federale in Lausanne and Vanderbilt University are collaborating in a broad materials science program at the FEL, that has recently produced first interesting results. One objective is the use of nonlinear optical spectroscopy to study artificial quantum structures. The first test of nonlinear spectroscopy actually removed one of the oldest gaps in this field, by measuring the two-photon absorption of the indirect gap of germanium, and providing after 18 years a positive test of the bassani-hassan theory. The details of the experiment clearly demonstrate that nonlinear optical spectroscopy is possible with the FEL. We will discuss the future plans in this direction, including additional results on other semiconductors. We will also report on the progress of the other parts of this multifaceted program, with particular emphasis on multiphoton photoemission, surface photovoltage studies and surface infrared absorption. In each case, we will show that the FEL's superior characteristics make it possible to solve open problems in semiconductor interface research, such as the role of strong fermi-level pinning and the details of the local electronic structure. Finally, we will comment on the general framework of this approach, which couples the FEL experiments and a network of other advanced facilities for the study of interfaces. We note the complementarity and mutual enhancement of the FEL-based techniques and of laboratory-based internal photoemission at Lausanne plus the extensive use of the novel technique of photoemission and x-rays at Lausanne, Wisconsin and Trieste.

*Work performed in full collaboration with J. T. McKinley, R. G. Albridge, A. V. Barnes, A. Ueda, X. Yang and N. H. Tolk, *Department of Physics and Astronomy, Vanderbilt University, Nashville, TN 37235,* and E. Tuncel, J. L. Staeli, and C. Coluzza, *Ecole Polytechnique Fédérale, CH-1015 Lausanne, Switzerland.*

Probing Terahertz Dynamics in Semiconductor Nanostructures with the UCSB Free-electron Lasers

S. J. Allen

Center for Free-electron Laser Studies
UC Santa Barbara, CA 93106

The UCSB free-electron lasers provide kilowatts of continuously tunable radiation starting from 120 GHz, the upper limits of conventional microwave electronics, and extending up to 4.8 THz. They have the potential to impact non-linear dynamics in semiconductor nano-structures, define the high frequency limits of conventional electronics and explore new phenomena that may enable new terahertz science and technology. We will review the following research activities:

- Terahertz relaxation of high speed resonant tunneling diodes.
- Photon assisted sequential resonant tunneling.
- Saturation spectroscopy .
- Resonant harmonic generation.

Saturation Spectroscopy of Free Carriers in Quasi-Two-Dimensional Systems

B. D. McCombe,# W. J. Li,# J. P. Kaminski,* S. J. Allen, Jr.* M. I. Stockman,+L. S. Muratov,+ L. N. Pandey,+ T. F. George,+ R. Ranganathan,** and W. J. Schaff++

 # SUNY Buffalo, Buffalo, NY;
* U.C.S.B, Santa Barbara, CA;
+ Washington State Univ., Pullman, WA;
** California State Univ., Northridge, CA;
+ + Cornell Univ., lthaca NY.

Far infrared non-linear (saturation) magneto-absorption experiments were carried out on a series of modulation doped single hetero-structure, multiple isolated-quantum-well (IQW) and coupled double-quantum-well (CDQW) structures, with doping density from 3×10^{10} cm^{-2} to 1.2×10^{11} cm^{-2}. Saturation of resonant absorption of free carriers in magnetic field (cyclotron resonance) was achieved with the Free Electron Laser (FEL) at UC Santa Barbara, which is tunable in the range of 63 μm to 2 mm, and is capable of peak power up to 40 KW. For magnetic field perpendicular to the plane of the QWs, large saturation intensities ($\sim$ KW/cm^2) were obtained. The population decay time of Landau levels (LL) is of the order of picoseconds from a 3-level model calculation.[1] This short lifetime is attributed to a rather uniform LL spacing and an Auger process induced by electron-electron scattering combined with emission of LO-phonons in the cooling processes. For magnetic field tilted away from the QW axis, anti-level crossing between LL and the confinement subband not only introduces non-uniform LL spacings but also suppresses the Auger process.[2] In this case, much lower laser intensities ($\sim$W/cm2) were needed to achieve saturation. The population decay time of the coupled LL-subband system is much longer ($\sim$ nsec). There is no apparent carrier density dependence of the decay time.

[1] R. Ranganathan, J. Kaminski, W. J. Li, J.-P. Cheng, and B. D. McCombe, Proceedings of the SPIE, 1678, 120 (1992)

[2] W. J. Li, B. D. McCombe, J. P. Kaminski, S. J. Allen, M. I. Stockman, L. S. Muratov, L. N. Pandey, T. F. George, and W. J. Schaff, 8th International Conference on Hot Carriers in Semiconductors, Oxford UK, August 16, 1993.

Application of Free-Electron Laser Technology to the Study of Energy Transfer in Disordered Solids

P.C Taylor and Z.V. Valdeny

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University of Utah
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Research on energy transfer in disordered solids at the University of Utah falls into four categories: (1) energy transfer mechanisms in disordered semiconductors, (2) photochemical and photophysical processes, especially those occurring at interfaces and on surfaces, (3) energy transfer mechanisms in disordered insulators, and (4) ultrafast and non-linear effects in disordered solids. Most of the experimental studies involve the use of lasers that simulate the properties of infrared free electron lasers (FELs), but two specific studies - nonlinear optical (NLO) responses of conducting polymers and photoluminescence excitation (PLE) spectroscopies of disordered semiconductors involve the use of the FEL facilities at Duke University and Vanderbilt University, respectively. Examples will be briefly described of recent progress in the four areas defined above. The NLO studies of conducting polymers and PLE studies of disordered semiconductors will be discussed in more detail.

Two-photon absorption of single crystal polydiacetylene-*para*-toluene-sulfonate (PDA-pTS) was measured over the range 0.95 to 1.5 μm by frequency doubling the light from the FEL facility at Duke. The intensity dependence of the absorption was used to separate the one-photon and two-photon contributions and the frequency dependence of the two-photon absorption coefficient β was measured. The two-photon absorption spectrum for PDA-pTS exhibits two peaks near 1.85 and 2.35 eV. These two peaks are assigned to the transitions involving the 2^1A_g and m^1A_g states in the gap, respectively. These assignments solve a long-time controversy concerning the location of these levels with respect to the 1^1B_u excitonic state at 1.95 eV above the valence band edge and the conduction band edge at 2.45 eV. From these experiments the contribution to the third-order susceptibility, Im$\chi^{(3)}$ is estimated to be 1 x 10^{-8} esu for the m^1A_g absorption. The large value of this contribution is attributed to the enhancement of the two-photon absorption due to absorption through a midgap state near 1.2 eV in PDA-pTS.

PLE spectra were measured in thin films of hydrogenated amorphous silicon (a-Si:H) over the range 0.9 to 1.2 eV and in partially-ordered GaAs$_{0.81}$Sb$_{0.19}$ over the range 0.34 to 0.37 eV using the FEL at Vanderbilt. The PLE spectra in a-Si:H required frequency doubling of the fundamental FEL light. In a-Si:H the PLE spectra taken using the FEL demonstrate clearly that the major absorption mechanism is probably a two-step process that proceeds through the manifold of deep defects (silicon dangling bonds) in these films. Before these measurements it was commonly believed that such two step processes were unimportant in high-quality material. Recent PLE measurements performed at higher energies using a Ti-sapphire tunable source confirm the conclusions drawn from the FEL experiments.

Fullerene Solids:
From Properties to Practical Applications

N. J. Halas

Department of Electrical and Computer Engineering and the
Rice Quantum Institute
Rice University
6100 S. Main
Houston, TX 77251

Fullerene solids are in many ways the prototypical molecular solid or organic semiconductor. In our research, we have discovered several highly unusual and unique properties of fullerene solids, and their interactions with other molecular or atomic species. We have reported a series of optical experiments on fullerene thin films that have led to a new type of time-resolved spectroscopy ideal for the study of slow electronic relaxations in solids. In addition, we have demonstrated the successful growth of single crystal fullerene thin films, the facile diffusion of silver atoms into fullerene solids, the excimer laser-induced conductivity change in fullerene thin films, and a novel application of fullerenes as tips for a scanning tunneling microscope.

Simulations of Laser Drive Dynamics in Materials and Surfaces

Barbara Garrison

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The Pennsylvania State University
University Park, PA 16802

The goal of simulations of laser drive chemical and physical dynamics in materials and surfaces is pursued through a combination of formalism development, reactive chemical dynamics and IR photon absorption and vibrational relaxation processes on H terminated diamond surfaces. The formalism for the transition rates of two photon absorption and Raman processes from pulsed laser sources is developed such that it is possible to explicitly include phase coherence between Gaussian pump and probe lasers in the simulation of transition rates. The atomic simulations of reactive chemical dynamics, in the growth of diamond thin-films from hydrocarbon mixtures, include molecular dynamics (MD) simulations of individual microscopic reactions as well as dynamics of many simultaneously diffusing and reacting species on macroscopic time and length scales. The single IR photon absorption and vibrational relaxation dynamics of CH vibrations on H-diamond {111} surface is simulated so as i) to compare and quantify the simulated CH line-shapes line-shifts with experimental observations, and ii) to form a basis for MD simulations of two IR photon pump-probe processes for CH stretching and bending vibrations in condensed phase materials.

Distance Dependence of Electronic Energy Transfer between Donor and Acceptor Adlayers:
p-Terphenyl and 9,10-Diphenylanthracene

Steven M. George

Dept. of Chemistry and Biochemistry
Univ. of Colorado .
Boulder, Colorado 80309-0215

Investigations of energy transfer between adlayers on single-crystal surfaces provide a unique opportunity to explore electronic energy transfer in restricted geometries. In this study, laser induced fluorescence techniques and donor quantum yield measurements were used to examine the distance dependence of electronic energy transfer between donor and acceptor adlayers on $Al_2O_3(0001)$. The donor adlayer was p-terphenyl, the acceptor adlayer was 9,10-diphenylanthracene and n-butane was the variable spacer adlayer. The electronic energy transfer rates versus spacer thickness were determined at both 30 K and 85 K in ultra high vacuum. The butane spacer experiments showed that the donor energy transfer rate decreased with a $1/d^3$ dependence, where d is the thickness of the spacer adlayer. Given a Forster quantum mechanical or a Kuhn classical energy transfer mechanism with randomly oriented dipoles, a $1/d^3$ distance dependence is consistent with resonance electronic energy transfer from a two-dimensional donor adlayer to a three-dimensional array of acceptors. The spacer measurements yielded a critical transfer distance of $d_0 = 44$ Å ± 4 Å at 30 K and $d_0 = 33$ Å ± 6 Å at 85 K. The rate of donor-donor intralayer energy migration was also determined by measuring the electronic energy transfer rate versus donor coverage on the acceptor adlayer. The donor quantum yield measurements versus donor adlayer coverage were consistent with the spacer results and indicated that electronic energy migration does not occur within the p-terphenyl adlayer. These results versus spacer thickness and donor coverage reveal that electronic energy transfer in spatially confined geometries can be described using a modified Kuhn energy transfer mechanism.

Ultrafast Dynamics in Chemical and Biochemical Systems

John D. Simon* and John H. Weare

*Chemistry Department,
University of California, San Diego*

An objective of the MFEL program is the measurement and interpretation of the processes that take place in biochemical and complex materials systems. Our experimental program focuses on understanding the dynamics of condensed phase chemical and biochemical systems. Experiments using FEL radiation are planned for studying vibration relaxation and dephasing dynamics of substituted cyanobenzenes in solution, vibrational relaxation dynamics in the primary steps of the phytochrome photocycle (the pigment responsible for many morphogenic processes in plants), electronic and vibrational relaxation and dephasing in bacterial chlorophylls and photosynthetic reaction centers. We will use the FEL to obtain transient infrared spectra of reaction intermediates in biochemical systems. The FEL experiments will be complemented by time-resolved studies carried out at UCSD. Using femtosecond spectroscopy we have demonstrated that electronic dephasing dynamics of large solute and biological molecules can be studied using degenerate four wave mixing spectroscopy. Optically heterodyned Kerr spectroscopy is also being used to examine fundamental details of the structure and dynamics of electrolyte solutions. We have also used time-resolved polarization spectroscopies to study conformation motions and electronic structure changes in biochemical systems. Results from these studies and those proposed for future use of the FEL will be discussed in the talk.

*Speaker

Parallel Computers in Large Scale "First Principle" Simulations of Complex Materials

J. H. Weare* and John D. Simon

Chemistry Department,
University of California, San Diego

In order to fully interpret FEL data (say vibrational spectra in complex biological molecules), it is necessary to make a connection between the measured spectra and the interactions in the system. Because these interactions are so strong, for complex systems this connection must be made by direct simulation (e.g., simulation of the motion and calculation of the power spectrum). The development of "first principles" simulation methods has led to the solution of problems that would have been considered intractable even five or ten years ago. Yet the application of these methods to most biochemical and material problems is still prevented by their extreme complexity and large particle size. Our program addresses this problem.

In this talk I will discuss calculations of the electronic properties and dynamic behavior of clusters of second row elements with the particle sizes from 10—150. Aggregates in this size range have technologically important properties (e.g. catalytic specificity), that are very different from aggregates of molecular sizes or the bulk, Because there is no experimental structural information, it is necessary to develop methods to identify the lowest energy geometry which is presumed to be responsible for their remarkable properties. We do this by "dynamical" simulated annealing. This is easy to implement and effective, however, a very efficient solution to the electronic Schrödinger equation must be available. Because of the unpleasant size scaling of the present algorithms, vector machine calculations are prohibitively expensive. We are developing new algorithms for parallel computers which promise to greatly increase throughput. In this talk I will present our results for carbon and metal clusters, discuss the status of our implementation on parallel computers and discuss some directions that we are taking to improve the algorithm.

*Speaker

Materials Speakers

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Growth Program

October 28, Thursday

8:00 – 8:30 am	Registration
8:30 – 8:45 am	Welcome

SESSION I   **Chairperson: S. Pantelides**

8:45 am	**T. Heinz**	"Optical Studies of Surface Dynamics"
9:25 am	**E. Williams**	"Stability and Mobility of Structure on Surfaces"
10:05 am	**A. Zangwill**	"Kinetics of Epitaxial Growth
10:45 – 11:10 am	***** Coffee Break *****	
11:10 am	**E. Kaxiras**	"Surface Diffusion and Expitaxial Layer-by-Layer Growth"
11:50 am	**M. Tringides**	"Surface Diffusion Measurements in Non-Equilibrium Systems"
12:30 – 2:00 pm	***** *Lunch* *****	

SESSION II   **Chairperson: A. Voter**

2:00 pm	**R. Gomer**	"Surface Diffusion Studies Via the Fluctuation Method"
2:40 pm	**M. Lagally**	"Scanning Tunnelling Microscope Studies of Surface Diffusion"
3:20 pm	**S. C. Ying**	"Kramer's Rate Theory and Surface Diffusion"
4:00 - 4:30 pm	***** Coffee Break *****	
4:30 pm	**B. Garrison**	"Computer Simulations of Surface Diffusion and Reaction"
5:10 pm	**M. El-Batanouny**	"Dynamics and Structure of Surfaces Studied by He Scattering"
7:00 - 9:00 pm	***** Banquet *****	
9:00 - 10:00 pm	Poster Viewing	

Growth Program (continued)

October 29, Friday

SESSION III	**Chairperson: P. Nordlander**	
8:45 am	**J. Prybyla**	"Ultrafast Laser-Probing of Hot Electron Dynamics at Surfaces"
9:25 am	**J. C. Tully**	"Energy Transfer at Metal Surfaces: Molecular Dynamics with Electron Transfer"
10:05 am	**N. Tolk**	"Energy-Solid Interactions Leading to Desorption"
10:45 – 11:10 am	***** Coffee Break *****	
11:10 am	**S. George**	"Resonant Desorption Vibrational Spectroscopy and Vibrational Energy Dissipation in Surface Adlayers Using the Infrared Free Electron Laser"
11:50 am		Speakers To Be Announced
12:30 – 2:00 pm	***** Lunch *****	
Session IV	**Chairperson: A. Eguiluz**	
2:00 pm	**Y. Chabal**	"Adsorbate Vibrations at Semiconductor Surfaces"
2:40 pm	**P. Guyot-Sionnest**	"Infrared-Visible Sum-Frequency Generation as a Probe of Vibrational States and Dynamics of Adsorbates at Surfaces and Interfaces"
3:20 pm	**T. Reinecke**	"Vibrations on Reconstructed Metal Surfaces"
4:00 – 4:30 pm	***** Coffee Break *****	
4:30 pm	**J.J. Quinn**	"Higher Multipole Edge and Surface Modes of Two and Three Dimensional Electron Gas Systems"
5:10 pm	**E.W. Plummer**	"Collective Excitations in Thin Films and Interfaces"
8:00 - 10:00 pm		Discussion Session Discussion leaders: D. Mills and M. Lagally

October 30, Saturday 9:00 am   Vanderbilt University Free Electron Laser Tour

Growth Abstracts in Order of Presentation

Optical Studies of Surface Dynamics

T. Heinz

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Stability and Mobility of Structures on Surfaces*†

Ellen D. Williams

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As the fundamental unit in mass transport on surfaces, steps are crucial to both growth and etching, and to the stability of surface morphology (nanostructure). Diffraction and imaging techniques (LEED, STM, REM and LEEM) have been used to measure step structure and to follow the evolution of step structure on vicinal Si surfaces. By analyzing the statistical distribution of step positions and the time-dependent step motion under equilibrium conditions, both the energetics governing the structure of steps and the fundamental kinetic processes governing step motion have been determined. Modifications of the step structure have been observed during thermal processing, oxidative etching, growth and under electromigration. The kinetics of morphological transitions following thermal jumps can be described in terms of the fundamental energies and rates of step attachment/detachment determined from observations of individual steps. Such investigations have potential application for predicting the stability of nanostructures. Metastable structures are observed following oxidative etching or electromigration, suggesting the action of kinetic instabilities in step motion. The compatibility of the data with predictions based on the Schwoebel instability [1] *vs.* more recent models of chaotic step bunching [2] is discussed.

1. R. L. Schwoebel and E. J. Shipsey, J. Appl. Phys. 37, 3682 (1966).
2. D. Kandel and J. D. Weeks, Phys. Rev. Lett. 69, 3758 (1992).

Kinetics of Epitaxial Growth

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A review is presented of the present state of theoretical understanding of the phenomenon of epitaxial growth. Emphasis is placed on the interplay between and relative usefulness of (i) equilibrium total energy calculations; (ii) kinetic Monte Carlo calculations; and (iii) stochastic and deterministic equations of motion. The relevance of such studies to both metal and semiconductor systems is pointed out. I will argue that a synergy exists between efforts to elucidate the detailed microscopic mechanisms at work in particular systems and efforts to identify global features present in all epitaxial growth systems.

Surface Diffusion and Layer-by-Layer Growth

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Surface Diffusion Measurements in Non-Equilibrium Systems

M. Tringides

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The study of surface diffusion at finite coverage is highly complex because of the variety of configurations present (monomers, different size or shape islands etc.) Surface diffusion becomes highly non-linear and the diffusion coefficient coverage dependent, as a result of the adsorbate-adsorbate interactions, which are present in such systems. Non-equilibrium processes, where a system undergoes a symmetry change (i.e. ordering out of an initially disordered state, disordering out of an initially ordered state, adsorption on an initially clean surface) studied as a function of temperature can be used to clarify how the overlayer configuration affects the diffusion barrier.

Non-equilibrium constant coverage experiments will be described on O/W(110) after a temperature quench. Non-equilibrium constant flux experiments will be described on Ag/Si(111) at low coverage. Different activation energies are extracted depending on the configuration of the system. The difference can be accounted for by the interactions. These results show that the diffusion coefficients measured depend on the method of measurement.

Surface Diffusion Studies via the Fluctuation Method

R. Gomer

James Franck Institute and Department of Chemistry
The University of Chicago

A brief description of the fluctuation method will be followed by a discussion of some recent results, specifically the diffusion of oxygen and of CO on the Mo(110) surface and of hydrogen on the Re (1010) surface.

Scanning Tunneling Microscopy Studies of Surface Diffusion

Max G. Lagally

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Diffusion of atoms on surfaces can be explored with the scanning tunneling microscope in two ways: either in "real" time or in a freeze-and-look mode. In real time, which actually is more properly called "experimental" time, the tip is continuously scanned over the sample as the motion of individual entities such as vacancies, adatoms of small clusters is followed. The actual event can, of course, not be observed, but the time between events can be determined and from this a diffusion coefficient extracted. A consideration in such measurements is the high electric field at the sample due to the proximity of the tip, which may perturb the measurement. We present examples of vacancy motion in Si(001) and GaP(001) in which in the former the influence of the tip is negligible, while in the latter it is the overriding factor in controlling the motion. We present a theoretical analysis of the vacancy motion in Si(001) that suggests that the vacancy moves by a concerted motion of a dimer. In the freeze-and-look mode, the motion is allowed to occur and the morphology or distribution of atoms is subsequently frozen if for analysis. Such measurements can be done in analogy with those using field ion microscopy, i.e., following single entities. Work of others for Sb diffusion on Si(001) is reviewed in this context. Finally under proper conditions the density of islands formed during early stages of deposition is related to the diffusion coefficient. Examples for Ge and Si diffusion on Si(001) are shown. All of these methods have a parameter space in which they are most applicable; unfortunately so far no two methods have been compared on the same system.

Kramer's Rate Theory and Surface Diffusion

S. C. Ying

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A microscopic theory for the diffusive and vibrational motion of absorbed atoms on a solid surface is presented. We evaluate and analyze the dynamical structure factor. The quasi-elastic scattering peak is found to consist of mixed contributions from both diffusion and vibration. The diffusive contribution is analyzed in terms of jump rates and multijump probabilities. The vibrational peak in the dynamic structure factor indicates both a shift and broadening from anharmonic effects and frictional damping. Applied to Na on Cu(001) surface, the theory agrees quantitatively with recent experimental measurements.

Computer Simulations of Surface Diffusion and Reaction

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Dynamics and Structure of Surfaces Studied by He Scattering

M. El-Batanoumy

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Ultrafast Laser Probing of Hot Electron Dynamics at Surfaces

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Surface reactions induced by ultrafast lasers are most often driven by hot electrons. To develop a detailed understanding of such reactions it is essential that the ultrafast dynamics of hot electron generation and relaxation in the substrate be well understood. Recently it was shown that in metals the phenomena was not being treated accurately (Fann et al., Phys. Rev. Lett., **68**,2834 (1991)). In semiconductors, hot electron relaxation can be quite complex as evidenced by the numerous optical studies of GaAs and GaAs-based quantum wells.

While ultrafast optical techniques have been applied extensively to the study of hot electron dynamics in GaAs, there have been very few such studies of Si. This is because conventional optical pump-probe schemes are difficult to interpret for indirect gap materials. This is unfortunate since understanding hot electron dynamics is key to exploiting its properties in new devices, as well as in developing new advanced processing/analysis methods. Silicon provides an importantly different substrate for studying ultrafast surface reaction dynamics for comparison to the many studies done on metal substrates.

Here we report the first direct subpicosecond (150 fsec) time-resolved measurement of the energy relaxation of optically excited electrons in Si. This is accomplished in a laser pump-probe format by analyzing the *photoelectron* spectrum generated by an ultrashort uv probe pulse. By this method, we obtain directly the time-evolution of the photo-excited electron distribution function (bulk and interface) in Si with 150 fsec resolution. These measurements would not be possible by the conventional reflectivity/transmission or luminescence schemes.

We have observed an electron-electron thermalization time of <120 fsec, an extremely fast *initial* electron cooling rate in the conduction band (cb), followed by an electron-phonon thermalization time of ~1 psec, and overall a very strong energy dependence to the electron scattering rate. At longer delays, the spectra give direct evidence for transient cb electron accumulation at the surface.

Our findings on Si(100) are considered in terms of their unique potential for driving surface chemical processes.

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Energy Transfer at Metal Surfaces: Molecular Dynamics with Electronic Transitions

J. C. Tully and M. Head-Gordon

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"Molecular Dynamics", the simulation, on the computer, of the detailed motions of an assembly of interacting atoms, is based on the Born-Oppenheimer approximation: the electron distribution adjusts adiabatically to the motion of the atoms. This approximation breaks down for adsorbate motion at metal surfaces, where the presence of metallic conduction electrons introduces the possibility of electron-hole (e-h) pair excitations or de-excitations. We have developed a method which treats atomic motion and electronic transitions self-consistently. E-h pair transitions are described by fluctuating forces and frictions, yielding a tractable molecular dynamics method that can be applied in full dimensionality. Most importantly, explicit expressions are obtained for the frictions and fluctuating forces that can be computed by ab initio methods.

The theory has been applied to carbon monoxide on the 100 surface of copper, demonstrating that e-h pair excitations have dramatic consequences. The simplest application is to nonradiative lifetimes of excited vibrations of the adsorbed CO molecule. Lifetimes of 3.3, 82, 2.3 and 108 ps. are calculated for the internal CO stretch, the CO - surface stretch, the frustrated rotation and the frustrated in-plane translation, respectively. These calculations are completely a priori, and are in good accord with the best current experimental estimates. In the absence of electronic friction, the calculated 0K lifetimes due to phonon excitations alone are greater than 100 ps. for all modes.

The influence of electronic transitions on sticking is found to be minor but not negligible. E-h pair transitions dominate the vibrational excitation or de-excitation of scattered molecules, and provide a convincing explanation for the observed trends in the vibrational excitation of nitric oxide scattered from silver. In addition, our simulations accurately reproduce recent observations of desorption induced by non-equilibrium heating of metallic electrons using femtosecond laser pulses.

Nonthermal Energy-Solid Interactions Leading to Desorption and Growth at Surfaces†

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We utilize a variety of photon, electron, and ion sources including the unique capabilities of the Vanderbilt Free-Electron Laser (FEL) to elucidate and resonantly enhance growth, desorption and cluster formation processes on insulator and semiconductor surfaces. The role of defects created by photon and electron irradiation leading to surface metalization is particularly emphasized in understanding desorption mechanisms. In addition, our experience with selective resonant processes at surfaces suggests that specific laser wavelengths profoundly affect growth processes.   It is recognized that early stages of growth on surfaces involve a complicated interplay of free radical adsorption, atomic and molecular surface diffusion and surface defects. Clusters can form with peculiar quantum mechanical properties that are not directly observed on either a macroscopic or atomic scale, but dominate fundamental macroscopic dynamical phenomena.   Emphasis is this talk will be on (a) first measurements of strong FEL wavelength-dependent damage thresholds on CVD-grown diamond films near the C-H stretch mode which under certain circumstances results in an annular region of new diamond growth which appears concentrically about the ablated crater; and (b) application of an optical-absorption technique during irradiation induced desorption to measure defect and cluster concentrations in real time. These experiments give strong evidence that desorption and growth processes may be initiated and controlled by the judicious application of wavelength and intensity dependent FEL pulses.

*Work performed in full collaboration with J. T. Mckinley, R. G. Albridge, A. V. Barnes, A. Ueda, S. Vijayalakshmi, D. Liu, J. Xu, N. Seifert, and J. L. Davidson, *Department of Physics and Astronomy, Vanderbilt University, Nashville, TN 37235* , A. Burger and M. George, *Department of Physics, Fisk University, Nashville, TN 37208,* and G. Margaritondo, *Ecole Polytechnique Fédérale, CH-1015 Lausanne, Switzerland,*

Non-Adiabatic processes in DIMET and Excitation in the STM Bistable Switch

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We discuss two perceived examples of nonadiabatic processes, desorption by intense femtosecond laser pulses' (DIMET) and excitation of an atom trapped in the double well between the surface and an STM tip2. In each case a model of a massive degree of freedom coupled to an electron gas out of equilibrium, in one case via a time-varying temperature and the other via a current, is appropriate. A path integral approach simplifies the model to motion of the massive particle in the presence of a generalized friction, plus a related stochastic force. Results are compared with experimental data.

References

1. D.M. Newns, T.F. Heinz, and J.A. Misewich, Progr. Theor. Physics, Kyoto,**106**, 411 (1991); T.F. Heinz, J.A. Misewich, and D.M. Newns., Phys. Rev. Lett., **68**, 3737 (1992).

2. S. Gao, M. Persson and B.l. Lundqvist, Solid State Commun. **84**, 271 (1992); R.E. Walkup, D.M. Newns and Ph. Avouris, Phys. Rev. **B48**, 1858 (1993).

Resonant Desorption Vibrational Spectroscopy and Vibrational Energy Dissipation in Surface Adlayers Using the Infrared Free Electron Laser

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Infrared free electron lasers provide a unique light source for surface spectroscopic and dynamics studies because of their wavelength tunability and high energy output. We are currently building a ultrahigh vacuum chamber for vibrational spectroscopic and dynamics experiments on the Mark III IRFEL at Duke University. This talk will cover the potential applications of the IRFEL for studies of vibrational energy relaxation and resonant desorption vibrational spectroscopy. Earlier results will be reviewed and future investigations will be discussed.

Adsorbate Vibrations at Semiconductor Surfaces

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Adsorbate vibrations are useful to determine surface structures and adsorbate bonding configurations. Critical to this analysis is the understanding of the **dynamical** interactions among various adsorbates, such as dipole, steric and subsurface interactions.

We consider these various dynamical interactions for the unusual hydrogen bonding arrangement found on vicinal Si(111) prepared by wet chemical processing. For surfaces miscut in the $<1\bar{1}2>$ direction, the double-layer steps are terminated by a dihydride. The unusual aspect is the strong steric interaction present between this dihydride and the terrace monohydrides below, resulting in large frequency shifts. Infrared absorption, Raman scattering and Sum-frequency Generation are used, along with ab-initio cluster calculations, to characterize this strained structure.

The clear separation of the various spectral modes makes it possible to identify and quantify the phenomenon of **vibrational energy diffusion** at surfaces, using two-color pump/probe techniques with SFG detection.

Vibrations on Reconstructed Metal Surfaces

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Higher Multipole Edge and Surface Modes of Two and Three Dimensional Electron Gas Systems

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and
[2]*Oak Ridge National naboratory*

When the electron density at the edge of a two dimensional electron gas (2DEG) decreases smoothly from its bulk value to zero, in andition to the regular edge plasmon, novel higher multipole edqe modes are found. It is not difficult to demonstrate that these modes are bulk 2D plasmons of the low density edge region that are unable to propagate into the bulk. Understanding these edge modes helps to clarify the nature on the higher multipole surface modes of a 3DEG. For degenerate semiconductors all of these excitations are in the infrared and their frequencies depend strongly on applied magnetic field.

Collective Excitations in Thin Films and Interfaces

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Growth Speakers

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